PREDICTION OF THE ELASTIC MODULUS OF PARTICULATE COMPOSITES BY MEANS OF A GENERAL EVALUATION METHOD USING A FOUR COMPONENTS MODEL

By

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Abstract

In this study, the elastic modulus of particulate composites by means of a model cube-within-cube is evaluated. The R.V.E. (representative volume element) is consisted by four components, of which one component corresponds to the filler and the rest three components correspond to the matrix. Because of the different composite geometry in the three directions, 3 submodels are created, one in each direction, providing 3 different values for the elastic modulus. The evaluation procedure considers triaxial stress situation in each part of the model and the elastic modulus is evaluated by means of the governing stresses and elongations equations of the model. The theoretical results are compared with those derived by existing equations in the literature as well as with experimental tensile results and ultrasonic measurements in epoxy / iron particulate composites.

Introduction

Addition of metal particles to polymers can produce a number of desirable effects, for example an increase in elastic modulus, a reduction in the coefficient of thermal expansion, an increase in electrical conductivity, an improvement in creep resistance and fracture toughness and an increase in density. However the fracture stress, strain and energy are decreased by the addition of rigid particles into the polymer matrix.

It must be noted that two other properties are of considerable importance in the processing and application of these materials. Firstly, the viscosity of the uncured mixture must be sufficiently low to permit the evacuation of air bubbles. Secondly, for many uses the glass transition temperature is critical. For example in high and medium voltage insulators the filled material must have a glass transition temperature of at least $120^{\circ}C$ in order to withstand the temperature attained around the electrode.

However, one of the main problem remains the prediction of the composite properties, when the properties of the constituent materials are known. The various theoretical models that have been proposed [1-21] to predict the mechanical properties of composites have emphasized particular parameters. The filler-volume fraction and the mode of packing were the parameters studied in the models presented in Refs [1-3], while the importance of the particle size on the final properties of the composites was discussed in Ref., (4-6,10,12-13]. The effect of the filler matrix adhesion on the mechanical behaviour of composites has been discussed in a series of models presented in Refs [7-11,13,15,17-21].

In this paper according to ref. [22] the elastic modulus of particulate composites is evaluated by means of a four parts model in cube-within-cube formation. Because of the different geometry of the model in each direction an anisotropy is appeared for the composite. Thus three different values are obtained for the elastic modulus (E_c), one in each direction. The lower values of E_c are close to tensile experimental results in epoxy / iron particulate composites while the higher values are close to ultrasonic measurements in the above composite for high filler content.

Theoretical Considerations

The theoretical analysis is based on the following assumptions.

i) The matrix and filler are elastic, isotropic and homogeneous.

ii) There is perfect adhesion between matrix and filler.

iii) The deformations applied to the composite are small to maintain linearity of stress-strain relations. From Fig. (1) for the filler volume fraction we have

$$\upsilon_f = a^3 / c^3 \tag{1}$$

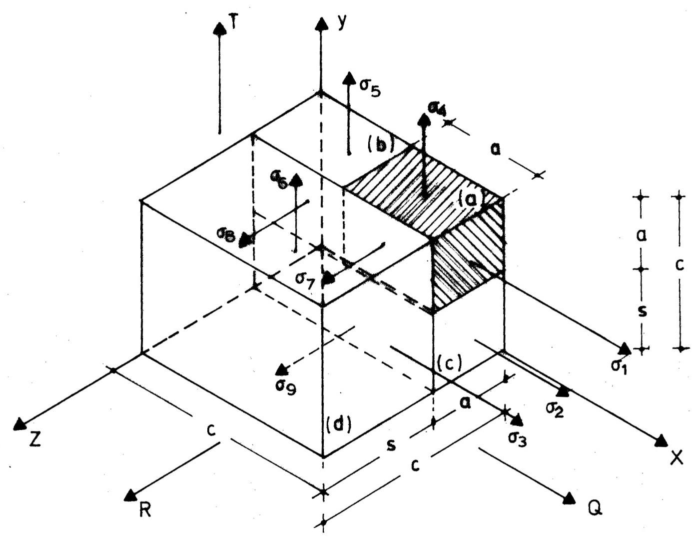

Fig. 1. The four parts model in cube-within-cube formation with triaxial stress situation in each component, for external stresses Q, T, R .

According to ref. [22] for external stresses Q, T and R in x, y and z directions respectively, the stress situation is assumed triaxial for each component of the composite as in fig. (1).

The equilibrium equations are given by

$$\sigma_1 \upsilon_f^{2/3} + \sigma_5\left(\upsilon_f^{1/3} - \upsilon_f^{2/3}\right) + \sigma_3\left(1 - \upsilon_f^{1/3}\right) = Q \tag{2}$$

$$\sigma_4 \upsilon_f^{2/3} + \sigma_5\left(\upsilon_f^{1/3} - \upsilon_f^{2/3}\right) + \sigma_6\left(1 - \upsilon_f^{1/3}\right) = T \tag{3}$$

$$\sigma_7 \upsilon_f^{1/3} + \sigma_8\left(\upsilon_f^{1/3} - \upsilon_f^{2/3}\right) + \sigma_9\left(1 - \upsilon_f^{1/3}\right) = R \tag{4}$$

To maintain the model geometry, the elongations equality in the three directions gives the following equations

$$\upsilon_f^{1/3} \cdot \varepsilon_x^{(\alpha)} + \left(1 - \upsilon_f^{1/3}\right) \cdot \varepsilon_x^{(b)} = \varepsilon_x^{(c)} = \varepsilon_x^{(d)} \qquad (5),(6)$$

$$\varepsilon_y^{(a)} = \varepsilon_y^{(b)} \tag{7}$$

$$\upsilon_f^{1/3} \cdot \varepsilon_y^{(b)} + \left(1 - \upsilon_f^{1/3}\right) \cdot \varepsilon_y^{(c)} = \varepsilon_y^{(d)} \tag{8}$$

$$\varepsilon_z^{(a)} = \varepsilon_z^{(b)} = \varepsilon_z^{(c)} \qquad (9),(10)$$

where the indexes (a), (b), (c), (d) correspond to the components (a), (b), (c) and (d) of the composite respectively.

The constitutive equation of the components of the composite are given by the generalized Hooke's law.

$$[\varepsilon]=\begin{bmatrix} \frac{1}{E} & -\frac{\nu}{E} & -\frac{\nu}{E} \\ -\frac{\nu}{E} & \frac{1}{E} & -\frac{\nu}{E} \\ -\frac{\nu}{E} & -\frac{\nu}{E} & \frac{1}{E} \end{bmatrix}[\sigma] \tag{11}$$

The constitutive equations of the total composite are

$$\varepsilon_{cx} = \frac{1}{E_{cx}}\left(Q - T\nu_{xy} - R\nu_{xz}\right) \tag{12}$$

$$\varepsilon_{cy} = \frac{1}{E_{cy}}\left(T - Q\nu_{yx} - R\nu_{yz}\right) \tag{13}$$

$$\varepsilon_{cz} = \frac{1}{E_{cz}}\left(R - Q\nu_{zx} - T\nu_{zy}\right) \tag{14}$$

The eqns (2)~(10) consist a system of 9 equations with 9 unknowns, the stresses σ_1 to σ_9. After solving for the stresses for each filler content, the strains ε_{cx}, ε_{cy}, ε_{cz} are evaluated. Then the elastic moduli of composite from eqns (12), (13), (14) as in ref. [22] are obtained. Three different cases of external stresses a) $Q=1$, $T=R=0$, b) $T=1$, $Q=R=0$, c) $R=1$, $Q=T=0$ were assumed.

Results and Discussion

In Fig. (2) the elastic modulus E_c is plotted versus filler volume fraction. In this figure the theoretical values predicted by simple models in cube within cube formation [23], where the effect of the Poisson ratio was not taken into consideration are also presented, cuves (f), (b) and (d), corresponds to eqns (A_4), (A_5) and (A_6) respectively (Appendix).

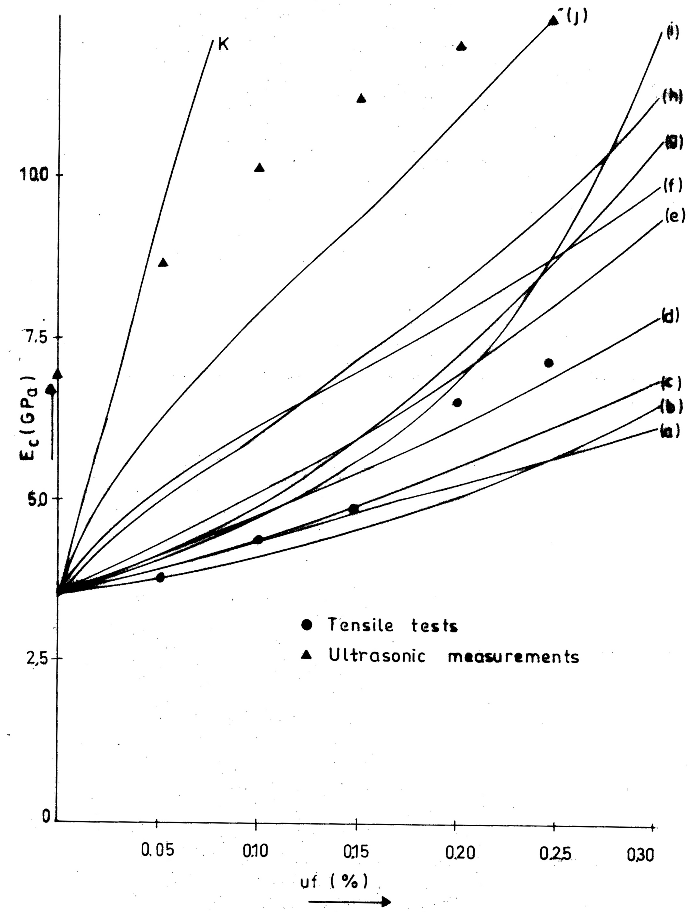

Fig. 2. Elastic modulus E_c versus filler volume fraction υ_f, in iron/epoxy particulate composites. (a) ; eqn (A_1), (b) ; eqn (A_5), (c) ; eqn (A_8), (d) ; eqn (A_6), (e); x direction of the presented model, (f); eqn (A_4), (g); eqn (A_2), (h); y direction of the presented model, (i); eqn (A_3), (j); z direction of the presented model, (k); eqn (A_7).

The curves (e) (x-direction), (h) (y-direction) and (j) (x-direction) correspond to the present submodels where the differences in Poisson ratio of two phases are taken into account. In the same figure the theoretical results predicted by eqns (A_1)~(A_3), (A_7) and (A_8), reported in Appendix, curves (a), (g), (i), (k) and (c) respectively also presented. It is observed in this figure that the values of E_c predicted by the simple models, in cube-within-cube formation, are closer to the experimental results in iron/epoxy particulate composites than those predicted by the presented submodels, in which the influence of the different Poisson ratios values in the two phases is taken into account. However the theoretical results predicted by the presented model in x direction are close to tensile experimental results, while the values predicted by the presented model in y direction are roughly accepted. The values of E_c predicted by the submodel in z direction are very high compared with the tensile experimental results. Therefore the appearance of such a model in the body of the composite seems to be possible when direction x corresponds to the load direction. However the discrepancies between the simple models in cube within cube formation and the presented submodels lead to the conclusion that, possibly one part of the transverse forces created by the different Poisson ratios in the two phases is eliminated by the interphase, which is a layer between the filler and matrix, having its own physicochemical properties which vary between those of the filler and those of the matrix. In other words the transverse stresses in each direction, possibly have to be multiplied by a coefficient k, $0 \le k \le 1$ because of the possible elimination of a past of them by the interphase. Although the provided values by the submodel in z direction are bounded by the theoretical results predicted by the Hashin and Shtrikman bounds, curves (k) and (c), however the predicted values by this submodel, for high filler content are close to the ultrasonic measurements, which for $\upsilon_f = 0{,}25$ give twice the value of E_c of the tensile experiment.

This fact gives rise to the assumption that probably there is an imperfect adhesion between the filler and the matrix in the used specimens which on the one hand can give noticeable differencies in the static measurements, while at the same time is not detected by the ultrasounds.

In fact the above mentioned discrepancies between the ultrasound method and static measurements appear also in the measurement of the Elastic modulus of the matrix only, where inverse phenomena can be observe with respect to molecular and intermolecular cohesion forces. Thus the small cohesion forces can not be noticeable by the static measurements but due to inertial phenomena and possibly small amplitude vibration can be noticeable by the ultrasound method. In the same figure we observe that eqns (A_1)~(A_3) and (A_8) give satisfactory approximation of the static measurement only for low filler volume fraction.

In Fig. (3) the reinforcing coefficient is plotted versus filler content. We observe that the reinforcing coefficient values predicted by the ultrasonic measurements are approximated satisfactorily by those predicted by the presented model in x direction, curve (d) and by the simple model in y direction, curve (c), which correspond to eqn (A_6).

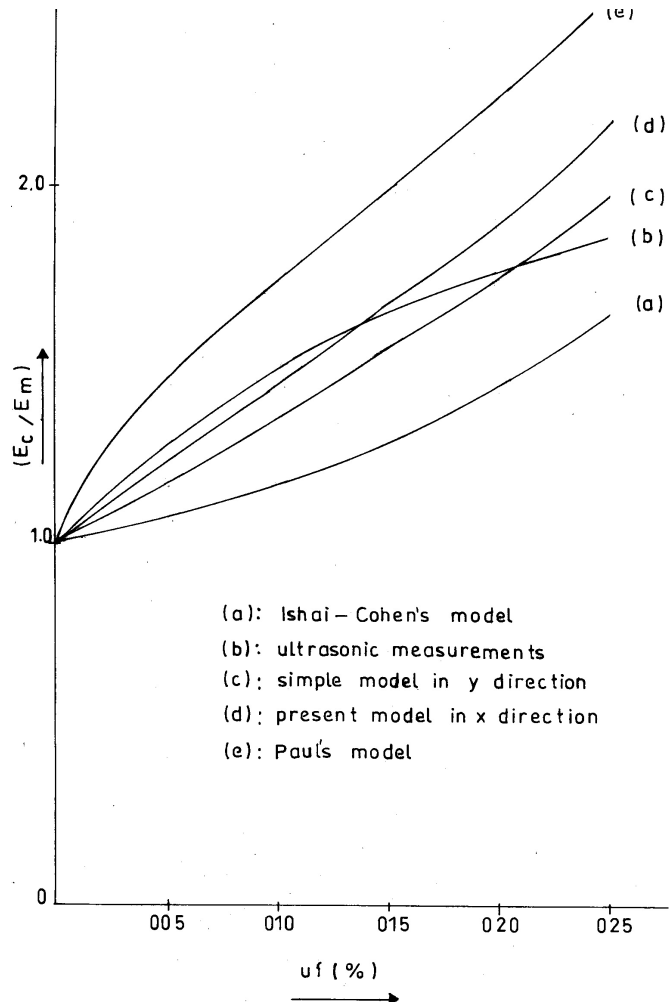

Fig. 3. Reinforcing coefficients in iron/epoxy particulate composites.

The measured high values in ultrasonic measurements are derived from ref. [31] and are supported from the results of refs [32, 33]. The computation of E_c by the ultrasound method is reported in ref. [31]. The tensile experimental results are derived from ref. [34]. For the computation of E_c the following values for the elastic constants of two phases were used

$$E_f = 210GPa \qquad E_m = 3{,}5GPa$$
$$\nu_f = 0{,}29 \qquad \nu_m = 0{,}36$$

Conclusions

The elastic modulus can be evaluated by the presented model when the orientation of the model is such that the x direction corresponds to the load direction.

It is possible that one part of the transverse forces in each direction, as they are considered in the present study, is eliminated by the interphase.

Because of inertial phenomena and small amplitude vibrations the measurements with the ultrasound method possibly do not detect some imperfections existing in the molecular and intermolecular cohesion and the imperfect adhesion between matrix and filler.

Although the ultrasonic measurements do not give the accurent values of the elastic modulus of the composites, they provide exactly its reinforcing coefficients.

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Appendix

The literature equations used for comparison are the followings

1. Einstein formula [24]

$$E_c = E_m\left(1 + 2{,}5\upsilon_f\right) \tag{A$_1$}$$

2. Guth and Smallwood equation [25,26]

$$E_c = E_m\left(1+2{,}5\upsilon_f + 14{,}1\upsilon_f^2\right) \quad (A_2)$$

3. Mooney equation [27]

$$E_c = \exp\frac{1+2{,}5\upsilon_f}{1-5\upsilon_f} \quad (A_3)$$

where for closed spheres packet $s = 1{,}35$.

4. Paul's model [28]

$$E_c = E_m\left(\frac{1+(m-1)\upsilon_f^{2/3}}{1+(m-1)\left(\upsilon_f^{2/3}-\upsilon_f\right)}\right) \quad (A_4)$$

where $m = E_f / E_m$.

5. Ishay-Cohen equation [29]

$$E_c = E_m\left(1+\frac{\upsilon_f}{\frac{m}{m-1}-\upsilon_f^{1/3}}\right) \quad (A_5)$$

where $m = E_f / E_m$.

6. Model 3 of ref. [23]

$$E_c = E_m\left(1+\frac{\upsilon_f}{\frac{m}{m-1}+\upsilon_f^{1/3}-\upsilon_f^{2/3}}\right) \quad (A_6)$$

where $m = E_f / E_m$.

7. Hashin and Shtrikman's bounds for E_c [30]. The upper and lower bounds are given respectively by

$$E_c = \frac{9\left(K_m + \dfrac{\upsilon_f}{\dfrac{1}{K_f - K_m} + \dfrac{3\upsilon_m}{3K_m + 4G_m}}\right)\left(G_m + \dfrac{\upsilon_f}{\dfrac{1}{G_f - G_m} + \dfrac{6(K_m + 2G_m)\upsilon_m}{5(3K_m + 4G_m)G_m}}\right)}{3\left(K_m + \dfrac{\upsilon_f}{\dfrac{1}{K_f - K_m} + \dfrac{3\upsilon_m}{3K_m + 4G_m}}\right) + \left(G_m + \dfrac{\upsilon_f}{\dfrac{1}{G_f - G_m} + \dfrac{6(K_m + 2G_m)\upsilon_m}{5(3K_m + 4G_m)G_m}}\right)} \qquad (A_7)$$

and

$$E_c = \frac{9\left(K_f + \dfrac{\upsilon_m}{\dfrac{1}{K_m - K_f} + \dfrac{3\upsilon_f}{3K_f + 4G_f}}\right)\left(G_f + \dfrac{\upsilon_m}{\dfrac{1}{G_m - G_f} + \dfrac{6(K_f + 2G_f)\upsilon_f}{5(3K_f + 4G_f)G_f}}\right)}{3\left(K_f + \dfrac{\upsilon_m}{\dfrac{1}{K_m - K_f} + \dfrac{3\upsilon_f}{3K_f + 4G_f}}\right) + \left(G_f + \dfrac{\upsilon_m}{\dfrac{1}{G_m - G_f} + \dfrac{6(K_f + 2G_f)\upsilon_f}{5(3K_f + 4G_f)G_f}}\right)} \qquad (A_8)$$

where K and G are the bulk and skew moduli.